PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of:		) Paper No. 7
APPLICANTS:	GEORGE E. MORRIS, ANDREW R. LUCY, WILLIAM A. GONG, MONICA C. REGALBUTO, & GEORGE A. HUFF JR.	)
SERIAL NO:	09/779287	) Group Art Unit:
FILED:	February 8, 2001	) Examiner: WALTER DEAN GRIFFIN
FOR:	PREPARATION OF COMPONENTS FOR TRANSPORTATION FUELS	) Attorney Docket No.: 37,248.04

DECLARATION UNDER 37 CFR § 1.131

Assistant Commissioner for Patents
Washington, DC 20231

ANY ADDITIONAL FEES REQUIRED CHARGE TO DEPOSIT ACCOUNT NO. 01-0528

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CERTIFICATE OF MAILING

I hereby certify that this correspondence is being deposited with the United States Postal Service with sufficient postage as first class mail in an envelope addressed to: Commissioner of Patents and Trademarks, Washington, D.C. 20231

on February 5 2003. By Carol M. Neth
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(Typed or Printed Name of Person Signing Certificate)

CONSIDERED
WG 4-10-2003

Examiner Griffin:

George Ernest Morris, Andrew Richard Lucy, William H. Gong, Monica C. Regalbuto and George A. Huff, Jr. hereby solemnly declare that:

1. They are the inventors who, on February 8, 2001, filed the above-identified application Serial No. 09/779,287.

2. They completed the invention, which is set forth in the above-identified application Serial No. 09/779,287, in the United States of America and the United Kingdom prior to September 28, 2000, the filing date of the application from which U.S. Patent No. 6,402,939 (Yen et al.) matured.

3. Prior to September 28, 2000, their invention as described and claimed in the subject application was completed in the United Kingdom, as evidenced by Laboratory Notebook 1396, pages 1 to 6, identified as EXHIBIT A, which illustrates the key points of declarants' process for oxidative desulfurization of a high boiling cut of a hydrodesulfurized diesel material. These pages memorialize and record experiments carried out in Hull, United Kingdom, a WTO member country, which serve to complete reduction to practice of the aforesaid invention.

4. To provide the higher boiling feedstock for the work at Hull, a hydrotreated refinery distillate was partitioned by distillation experiments carried out in United States under the supervision of Monica C. Regalbuto who is one of the undersigned declarants.

5. Under the supervision of Dr. George E. Morris, who is one of the undersigned declarants, Mr. Peter Anderson and Mr. Andrew Blanchard, Research Technicians, Hull Research Center, Hull, UK, conducted oxidation-simultaneous extraction experiments described in

a BP Chemicals Limited Laboratory Note Book 1396, pages 2 to 6. The work described shows that the sulfur species concentrated in a higher boiling cut of a hydrodesulfurized diesel material was oxidized with a soluble quaternary ammonium salt in an immiscible aqueous phase comprising hydrogen peroxide and phosphotungstic acid; separation from the reaction mixture of both an organic liquid and immiscible aqueous phase containing a portion of the oxidized sulfur species; and further removal of the oxidized sulfur species by silica sorption. The aqueous hydrogen peroxide phase and catalyst system can be reused with a fresh diesel charge. Comparative examples (not described in pages of the exhibit) showed that for the same reaction times and overall hydrogen peroxide usage greater amount of sulfur are treated and removed from the higher boiling cut than the corresponding full range material.

6. In this work Mr. Anderson charged a suitably equipped nitrogen purged glass reactor with S-25-288C-FBP (251.2 g), a.k.a. LS-98-25 (550°F FBP), aqueous hydrogen peroxide (61.6 g of 26.0 percent by weight), Aliquat® 336 (1.45 g) and an aqueous solution of phosphotungstic acid (0.83 g in 5.6 g water) and water (126.0 g). The hydrogen peroxide was equivalent to 8.2 percent by weight in the total aqueous phase. The reaction mixture was heated to 60° C. with stirring during 30 minutes and maintained at 60° C. with stirring during 4 hours. After cooling to ambient temperature the organic phase (248.9 g) was separated from the aqueous phase (189.3 g) and another, viscous brown oily phase. A sample of the organic phase was identified as PS-25-288C-FBP and retained for analysis which gave 43 ppm sulfur and 29 ppm nitrogen. The recovered aqueous phase contained 6.9 percent by weight hydrogen peroxide. The viscous brown oily phase was dissolved in methanol (40.0 g, recovered 42.0 g). Analysis of the methanol solution gave 30 ppm sulfur and 1100 ppm nitrogen.

7. Similar portions of PS-25-288C-FBP (total 186.1 g) were passed through one of two silica columns (11.9 g each). Analysis of

the product recovered (156.2 g) after silica treatment gave 0.3 ppm sulfur and 2.5 ppm nitrogen. A band of dark brown material was retained on the silica. The brown band was eluted from the column with methanol (23.0 g). On analysis it was found to contain 110 ppm sulfur and 58 ppm nitrogen.

8. Copies of the above referenced Laboratory Notebook pages are attached as Exhibits. The Exhibits are a true copy, except that the dates thereof and unrelated subject matter have been blanked-out, but all the dates are prior to September 28, 2000.

9. The undersigned declare further that at all statements made herein of their own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. 1001 and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

G E M — Date 28 JANUARY 2003

GEORGE ERNEST MORRIS

A. Lucy — Date 28th JANUARY 2003.

ANDREW RICHARD LUCY

William H. Gong — Date Feb. 3, 2003

WILLIAM H. GONG

Monica C. Regalbuto — Date Jan. 30, 2003

MONICA CRISTINA REGALBUTO

George A. Huff Jr. — Date Feb 3, 2003

GEORGE A. HUFF JR.

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BP CHEMICALS LIMITED

HULL RESEARCH CENTRE

LABORATORY NOTE BOOK

1396

OXIDATIVE DESULFURISATION OF DIESEL FUEL.

This work is a continuation of a series of experiments investigating reduction of sulphur concentration in diesel fuel using an oxidative process. Initial experiments are recorded in Laboratory note book 1292 pages 103 - 152.

Oxidative desulphurisation of diesel sample LS-98-25/ 550°F FBP 4hour reaction duration.

Date :

Ref : 1396/002

Aim This reaction is the first in a series of experiments in which we will evaluate the time dependancy of the rate of oxidative desulphurisation in a sample of diesel oil with fractional boiling point split > 550°F containing approx. 60 ppm sulphur. Data collected from this and other reactions in the series will be used to construct a mass balance showing the distribution of phase transfer agent, catalyst and sulphur.

Reaction charge.

	Required (g)	Actual (g)
Oil phase LS-98-25 (550°F FBP)	250.00	251.71
Aliquat 336®	1.45	1.45
Water	125.14	125.96
26% Hydrogen peroxide Soln	61.60	61.58
Phosphotungstic acid	0.81	0.827
in water	4.86	5.575

Cont.

P.J. Anderson

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All the listed materials were charged directly into a 2 ltr. reaction flask fitted with stirrer (~300 r.p.m) and overhead water cooled condenser. The contents of the reactor were heated to 60°C and held at that temperature for four hours.

Record of reaction temperature.

Time (H.m)	Elapsed time (m)	Temperature (°C) Contents	Skin
09:30	–	23	–
10:00	0	59	< 140°C
10:30	30	63	< 140°C
11:00	60	60	< 110°C
11:30	90	60	As above "
12:00	120	60	—
12:30	150	60	—
13:00	180	60	—
13:30	210	60	—
14:00	240	60	—

Reaction was shut down at 14:00 hr heater lowered and allowed to cool below 25°C before transfer to separation funnel.

Total mass of reaction product = 440.14 g

Liquid / Liquid Separation

Mass of separated Aqueous phase = 189.27 g

*Mass of heavy brown phase = 2.00 g

Mass of diesel phase = 248.87 g

* The heavy brown material was collected from the reaction flask and the separation funnel by first charging 40.0 g methanol into the reaction flask shaking to dissolve, then transferring into the separation funnel shaking to dissolve. A mass increase of 2 g was observed e.g. Solution wt. = 420 g,

Loading Silica absorption Columns.

	C1	C2
C3		
183.30 loaded with Silica	= 163.36	162.21
172.50 g Un loaded column mass	= 151.47 g	150.28
10.80 g Mass of Silica g	= 11.87	11.93
Not used. Col + Retained oil + Silica (g)	= 17806	175.50
Retained oil	= 1472	13.29

62.77g Sample of whole oxidative oil phase taken for Ref and analysis. Mass of diesel passed over Silica columns = 248.87g − 62.77 = 186.10g

1st column Mass of 1st fraction collected from base of Silica column = 93.24

2nd column Mass of 1st fraction collected from base of Silica column = 62.92g

Total = 156.16

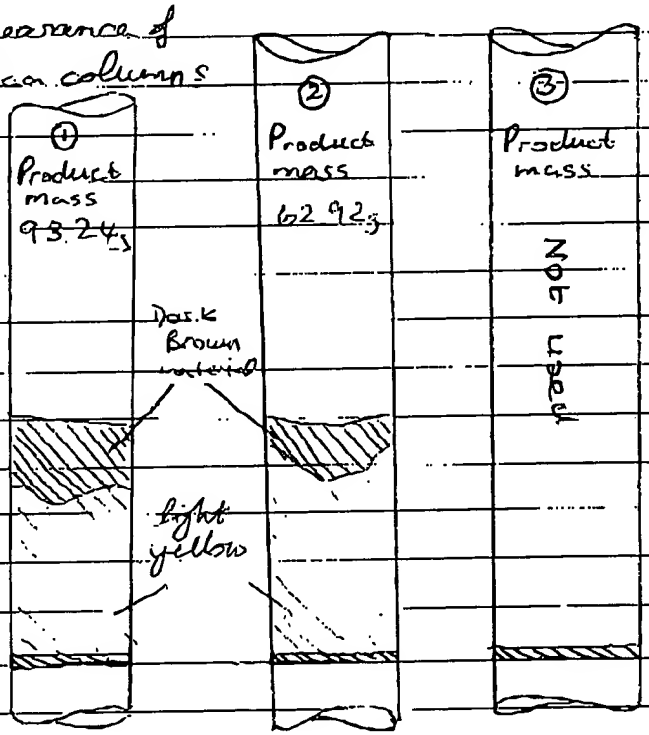
Retained mass of oil on columns = 28.01

Notes:

① Because of the apparent higher concentration of heavy brown material in the whole oxidative oil product the flow rate was greatly reduced. This reduction in flow rate was a result of a dense band of brown material which formed with in the top 1cm of the Silica column.

② Because of the low flow rate two additional Silica adsorption columns were set-up to process the remaining oil phase. Column 3 not used.

Appearance of Silica columns



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Analysis of aqueous phase for hydrogen peroxide

Dilution of aqueous phase

Mass of aqueous phase = 4.60 g
added water = 48.07 g

Dilution factor = 52.67 / 4.60 = 11.45

Analysis

	A	B	C
Sample wt. (g)	2.24	2.31	2.56
final titre vol. (cm^3)	41.00	49.60	47.20
last titre vol. (cm^3)	32.90	41.15	38.50
titre vol (cm^3)	8.10	8.45	8.70

Calculations

A $\frac{8.1 \times 0.1 \times 1.701}{2.24} \times 11.45 = 7.04\%$ wt.

B $\frac{8.45 \times 0.1 \times 1.701}{2.31} \times 11.45 = 7.12\%$ wt.

C $\frac{8.70 \times 0.1 \times 1.701}{2.56} \times 11.45 = 6.62\%$ wt.

Mean value 6.93 %wt hydrogen peroxide

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Methanol washing of t.o. 2 cm of silica adsorption columns

Both of the silica adsorption columns were washed with ~ 20g methanol to produce a solution of the brown material retained on the top two cm. of each column. This solution would be assessed for solid content and sent for Nitrogen, Sulphur and tungsten analysis.

Assessment of % solid in solutions of heavy brown oil and estimation of the actual mass of this material produced during the reactions.

1396-03

Solid content of heavy brown substance in methanol solution

Residual material washed from base of reaction flask and separating funnel

	1	2	3
Dish plus dried solid	1.9568	1.9242	2.0129
Dish plus solution	2.5583	2.5332	2.7312
Empty dish	1.9208	1.8871	1.9694
Wt. of solution	0.6375	0.6661	0.7618
Wt. of solid	0.036	0.0371	0.0435
% solid in solution	5.647059	5.569734	5.71016
Mean Value	5.642518		

Mass of methanol post washing (g)	42
-Mass of brown heavy oil (g)	2.269773

Solution obtained from washing the top two cm of the silica column with methanol

	1	2	3
Dish plus dried solid	2.6234	2.5924	2.938
Dish plus solution	3.4791	3.248	2.9724
Empty dish	2.8064	2.575	2.922
Wt. of solution	0.6727	0.673	0.6504
Wt. of solid	0.017	0.0174	0.016
% solid in solution	2.527129	2.585438	2.460025
Mean Value	2.524197		

Mass of methanol post washing (g)	23
-Mass of brown heavy oil (g)	0.580565

Total –mass of brown material (g)–	2.950339

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Note: In the first reaction of this series which evaluated the oxidative desulphurisation of the full range sample the mass of brown material produced was estimated to be 2.1g ~ 1/3 lower than that produced in this experiment conducted with the higher boiling point material.

P.S. Anderson